

Patent Application of
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and
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for

**TITLE: LAMINATED TUBE FOR THE TRANSPORT OF CHARGED PARTICLES
CONTAINED IN A GASEOUS MEDIUM**

CROSS-REFERNECE TO RELATED APPLICATIONS

This application claims the benefit of Provisional Patent Application Ser. Nr. 60/419,699, filed 2002, October 18. This application is related to Provisional Patent Application Ser. Nr. 60/210,877, filed June 9, 2000 now Patent Application Ser. Nr. 09/877,167, Filed June 8, 2001; Provisional Patent Application Ser. Nr. 60/384,864, filed June 1, 2002 now patent Application Ser. Nr. 10/449,344, Filed May 30, 2003; Provisional Patent Application Ser. Nr. 60/384,869, filed June 1, 2002 now patent Application Ser. Nr. 10/449,147, Filed May 31, 2003; Provisional Patent Application Ser. Nr. 60/410,653, filed September 13, 2002 now patent Application Ser. Nr. 10/661,842, filed September 12, 2003; and Provisional Patent Application Ser. Nr. 60/476,582, filed June 7, 2003.

FEDERALLY SPONSORED RESEARCH

The invention described herein was made with United States Government support under Grant Number: 1 R43 RR143396-1 from the Department of Health and

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Human Services. The U.S. Government may have certain rights to this invention.

SEQUENCE LISTING OR PROGRAM

Not applicable.

BACKGROUND-FIELD OF INVENTION

This invention relates to laminated capillaries which are used for interfacing higher pressure ionization sources to lower pressure ion destinations such as mass spectrometers, ion mobility spectrometers, and ion beam targets.

BACKGROUND-DESCRIPTION OF PRIOR ART

Dispersive sources of ions at or near atmospheric pressure, such as, atmospheric pressure discharge ionization, chemical ionization, photoionization, or matrix assisted laser desorption ionization, and electrospray ionization, generally have low sampling efficiency through conductance or transmission apertures and capillaries or tubes. Less than 1% [often less than 1 ion in 10,000] of the ion current emanating from the ion source is detected in the lower pressure regions of the present commercial interfaces for mass spectrometry.

Figure 1 show simulated trajectories of ions approaching a capillary entrance from a 400 V/mm ion source region into the relatively field-free inner channel of a capillary. A viscous [gas] flow velocity component is added to these ions in the direction of the capillary flow. This simulation shows the electric field penetration from the source region creates significant **dispersion of ions** and loss of ions to the walls at the inlet of a capillary. The losses of ions to walls will generally have two consequences; first, in the case of **conducting [metal] capillaries**, the ions will give up charge (usually through a redox process) or, second, in the case of **dielectric materials [glass]** the ion will accumulate on the surface and further retard introduction of subsequent ions into the flow through the capillary. Either way, the ions are primarily lost at or slightly downstream of the entrance of the capillary tube.

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U.S. patent 4,542,293 Fenn et al. (1985)¹ demonstrates the utility of utilizing a **dielectric [glass] capillary with metal ends with a large electric potential difference along the axis of the capillary, referred to as capacitive charging**, to transport gas-phase ions from atmospheric pressure to low pressure where the viscous forces within a capillary push the ions against a electrical potential gradient. This technology has the significant benefit of allowing grounded needles with electrospray sources. Unfortunately, this mainstream commercial technology² transmits only a fraction of a percent of typical atmospheric pressure generated ions into the vacuum. The majority of ions are lost at the inlet of the capillary due to the dispersive electric fields, at the inlet, dominating the motions of ions (Fig 1).

The requirement for **capacitive charging** of the dielectric tube for the transmission of ions, as well as, the acceptance or entry of ions into the capillary, is highly dependent on the charges populating the inner- and outer-surface of the capillary. This dependence of surface charging limits the acceptance and transmission efficiencies of Fenn et al.'s technology. Contamination of the large surface area of the inner-walls of the capillary from condensation, ion deposition, particulate material or droplets can change the surface properties and therefore reducing these efficiencies. In addition, since a large amount of energy is stored within the capillary, contamination can lead to electrical discharges and damage to the capillary, sometimes catastrophic. Therefore, care must also be taken to keep the inner- and outer-surfaces clean and unobstructed, presumably in order not to deplete the image current that flows on the outer-surface of the dielectric or the current that flows along the inner-surface.

Examples of metal capillaries are disclosed—for example, in U.S. patents 4,977,320 to Chowdhury et al. (1990)³, and 6,583,408 B2 (2003)⁴ and in U.S. patent application publication 2002/0185559 A1 (2002)⁵ both to Smith et al. Chowdhury et al. and Smith et al. both demonstrated the use of **heated metal capillaries** to both generate and transmit ions into the vacuum. The efficiencies of these devices are low as well. This technology samples both ions and charged droplets into the

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capillary where, with the addition of heat, ion desorption is facilitated. Drops undergoing coulomb explosions inside of a restricted volume of the lumen of the capillary will tend to cause dispersion losses to the walls were the charges are quickly neutralized and not resulting in the surface charging up. But similar to Fenn et al.'s dielectric capillary, this technique suffers the same limitation from losses at the inlet due to the dispersive electric fields (Fig 1), as described above.

Lin and Sunner (1994)⁶ studied a variety of effects on transmission through **tubes of glass, metal, and Teflon**. A wide variety of parameters were studied including capillary length, gas throughput, capillary diameter, and ion residence time. Effects from space charge, diffusion, gas flow, turbulence, spacing, and temperature were evaluated and discussed. Some important insights were reported with respect to general transmission characteristics of capillary inlets. However, they failed to identify field dispersion at the inlet as the first step in the loss of ions. In the case of glass capillaries, this dispersion and eventual impact of the ions on the inner-surfaces of capillary lumen leads to charging of the inner-surface of the capillary lumen at the entrance of the capillary preventing ions from entering into the capillary.

Several approaches have been proposed to eliminate or reduced the charging of the surfaces at the entry of **glass or dielectric capillaries**—for example, in U.S. patent 5,736,740⁷ (1998) and 5,747,799⁸ (1998) both to Franzen, U.S. patent 6,359,275 B1 to Bertsch et al. (2002)⁹; and U.S. patents 6,486,469 B1 (2002)¹⁰ and 6,583,407 B1 (2003)¹¹, and U.S. patent application publication 2003/003452 A1 (2003)¹² all to Fischer et al. Franzen (U.S. patent 5,736,740) proposes the use of a **highly resistive coating on the inner surfaces** of the capillary tube or use capillaries that are themselves highly resistive, such as, glass capillaries, to prevent charge accumulation as a means to facilitate the focusing of ions toward the axis of the capillary. Although it is difficult to distinguish this art from Fenn et al. (U.S. patent 4,542,293), in that the glass tubes in both approaches are highly resistive [or weakly conducting dielectrics], Franzen does argue effectively for the need to control the inner surface properties and therefore the internal electric fields. Irregardless,

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Franzen's approach will suffer from the same limitations as Fenn's, that is loss of ions in the dispersive electric fields at the inlets of capillaries and apertures.

Bertsch et al. (6,359,275 B1) proposes a similar approach to Franzen to prevent charging of the surface by coating the inner-surface. But unlike Franzen, Bertsch et al. coats the inner-surface of the capillary near the capillary entrance with a **conductive material**, thereby bleeding away any charge that builds up on the inner-surface to the end-cap. Bertsch et al. eliminates surface charging while still keeping the benefits of the dielectric tube transport in the nondispersive region [downstream region] of the capillary. This approach addresses the problem of charge accumulation on the inner-surfaces, but it does not remove the significant losses of ions at the inlet due to dispersion (Fig 1). Again, suffering the same limitations of Fenn et al.'s, Franzen's, and Chowdhury et al.'s devices—lose of ions at the inlet due to dispersive electric fields.

Franzen (U.S. patent 5,747,799) and W.O. patent 03/010794 A2 to Forssmann et al. (2002)¹³ addresses the need to focus ions at or into the **inlet of capillaries and apertures** in order enhance collection efficiencies by the use of a series of electrostatic lens at or in front of the inlet. In Franzen's device the ions are said to be first, attracted to the inlet by **electrostatic potentials** and once in the vicinity of the inlet the ions are entrained into the gas flowing into the tube or aperture by **viscous friction**. This invention fails to account for the dominance of the electric field on the motion of ions in the entrance region. At typical flow velocities at the entrance of tubes or apertures, the electric fields will dominate the motion of the ions and the ions that are not near the capillary axis will tend to disperse and be lost on the walls of the capillary or aperture inlet. With this device, a higher ion population can be presented to the conductance opening at the expense of higher field ratios across the aperture or along the capillary but at the expense of higher dispersion losses inside the aperture or tube.

Forssmann et al. (03/010794 A2) describes a series of electrodes, **or funnel**

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optics, upstream of the capillary inlet in order to concentrate and direct ions toward or into the capillary inlet. This approach utilizes funnel optics in front of an electrospray source in order to concentrate ions on an axis of flow by imposing focusing electrodes of higher electrical potential than the bottom of the so called accelerator device, the first electrode in the series. This device frankly will not work. The ions formed by the electrospray process will be repelled by this funnel optics configuration and little to no transmission of ions to the aperture or capillary inlet will occur. Most of the inertial energy acquired by the ions in the source region is lost to collisions with neutral gas molecules at atmospheric pressure; consequently the only energy driving the ions in the direction of the capillary inlet or aperture will be the gas flow which under normal gas flows would be insufficient to push the ions up the field gradient imposed by the funnel optics. This device does not operate in fully developed flow as will be described in the present invention.

U.S. patents 6,486,469 B1 (2002) and 6,583,407 B1 (2003); and U.S. patent application publication 2003/003452 A1 (2003) to Fischer et al. all utilize **external electrodes** and butted **dielectric tubes** to provide enhanced control of the **electric fields within** the capillary. While Fischer et al. (6,583,407 B1) utilize the conductive coating proposed by Bertsch et al. (6,359,275 B1) to eliminate surface charging, all three devices do not address issues related to inlet losses due to dispersive electric fields at the inlets of capillaries and apertures, as presented in Figure 1. In addition, all these devices still utilize significantly large dielectric inner-surfaces with the associated problems with surface charging, contamination, and discharge.

REFERENCES

- ¹ Fenn, J.B., Yamashita, M., Whitehouse, C., "Process and apparatus for changing the energy of charged particles contained in a gaseous medium," U.S. Patent 4,542,293 (September 17, 1985).
- ² See Analytica of Branford, Branford, CT, <http://aob.com>; Agilent Technologies,

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- ³ Chowdhury, S.K., Katta, V., Chait, B.T., "Electrospray ionization mass spectrometer with new features", U.S. Patent 4,977,320 (December 11, 1990).
- ⁴ Smith, R.D., Kim, T., Tang, K., Udseth, H.R., "Ionization source utilizing a jet disturber in combination with an ion funnel and method of operation," U.S. Patent 6,583,408 B2 (June 24, 2003).
- ⁵ Smith, R.D., Kim, T., Udseth, H.R., "Ionization source utilizing a multi-capillary inlet and method of operation," U.S. Patent Application Publication 2002/0185595 A1 (December 12, 2002).
- ⁶ Lin, B., Sunner, J., "Ion transport by viscous gas flow through capillaries", J. Am. Soc. Mass Spectrom. 5, pages 873-885 (1994).
- ⁷ Franzen, J., "Method and device for transport of ions in a gas through a capillary," U.S. Patent 5,736,740 (April 7, 1998).
- ⁸ Franzen, J., "Method and device for the introduction of ions into the gas stream of an aperture to a mass spectrometer," U.S. Patent 5,747,799 (May 5, 1998).
- ⁹ Bertsch, J.L., Fisher, S.M., Riccomini, J.B., "Dielectric conduit with end electrodes", U.S. Patent 6,359,275 B1 (March 19, 2002).
- ¹⁰ Fisher, S.M., Russ, C.W., "Dielectric capillary high pass ion filter", U.S. Patent 6,486,469 B1 (November 26, 2002).
- ¹¹ Fisher, S.M., Russ, C.W., "Method and apparatus for selective ion delivery using ion polarity independent control", U.S. Patent 6,583,407 B1 (June 24, 2003).
- ¹² Fisher, S.M., Russ, C.W., "Dielectric capillary high pass ion filter", U.S. Patent Application Publication 2003/0034452 A1 (February 20, 2003).
- ¹³ Forssmann, W-G, John, H., Walden, M., "Mass Spectrometry Device," WO Patent 03/010794 A2 (February 6, 2003).

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SUMMARY OF INVENTION

In accordance with the present invention a stratified or laminated tube comprises alternating layers of conducting electrodes and insulating (or dielectric) bases with an inlet for the introduction of gas-phase ions or charged particles and an exit, with an optional high-transmission surface populated with a plurality of openings upstream of stratified tube and a counter-flow of gas, for the introduction of ions into a lower pressure region. The electric potentials applied to the conducting electrodes are intended to provide a potential surface for the transfer of substantially all ions from an ion source region into the inlet of the laminated tube, transfer through the tube with minimal losses, and introduce the ions into a lower pressure region of user definable initial and exit potentials relative other components in the device. In some modes of operation the potentials can be varied to select specific species based on mobility.

To avoid the dispersion losses characterized by Fig 1 the current device eliminates the field penetration into the capillary tube from a higher field source (or optics region) upstream from the inlet of the capillary tube by applying accelerating potentials to successive layers of the laminated tube. The ions entering the tube are accelerated through the **Region of Flow Development** as illustrated in Fig 2 by these successive laminates. Since the velocity of the gas velocity at the entrance of the tube (v_{ent}) is substantially below the maximum velocity (v_{max}) in fully developed flow, the current device delays the dispersion (if at all) until the viscous forces are more capable of overcoming the dispersive effects from decreased electric fields. Once the ions traverse this Region of Flow Development, dispersive electric fields can be applied through successive laminate potentials in order to accomplish a variety of controlled optical processes; namely, to allow the ions to traverse a repulsive gradient, to select specific ions based on ion mobility, to store ions for brief periods, and to focus ions.

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Delaying dispersion until fully developed flow exists will eliminate the significant losses that occur at the entrance to the capillary. And in addition, by delaying dispersion until the ions are in fully developed gas flow the motion of the ions will be dominated by viscous forces in the controlled electric fields within the tube. Fig 3 shows a graph of the **Entrance Boundary Distance (L_{ent})** as a function of tube diameter showing the requirement to delay dispersion for many centimeters [down the length of the tube] with larger diameter tubing while reducing this distance to below a millimeter in smaller diameter tubing—illustrating the dimensional requirements for the current device. The dimensional requirements for the present device also indicate the need for macro- and possible micro-fabrication processes to create precision laminates.

Figs 4A thru 4D show computer simulation of the operation of the present laminated tube device with A) a uniform electric field generated through the entire length of the capillary (Note that dispersion is delayed in this embodiment until the exit of the tube), B) a dispersive well created by applied DC potentials located in a region of fully developed flow (Note a lower electric field at the exit of the tube allows higher inlet fields), and C) a focusing region created by applied RF potentials located in a region of fully developed flow (Note that RF potentials can be used to overcome diffusion losses in long tubes once the ions have traversed L_{ent}).

OBJECTIVES AND ADVANTAGES

Accordingly, besides the objects and advantages of the laminated high transmission surfaces described in our co-pending patents, several objects and advantages of the present invention are:

(a) to provide a laminated tube with **no or minimal losses** of ions or charged particles while transferring the ions into a lower pressure regions for mass spectrometric analysis, ion mobility analysis, and or ion beam deposition or ion chemistry,

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(b) to provide a laminated tube that substantially all the ions are transfer through the tube and are not deposited and **contaminating the lumen of the tube**, leading to a buildup of surface charged,

(c) to provide a laminated tube the restricts the flow of gas into the lower pressure regions, thereby **reducing the gas-load** on the device and any vacuum pumping associated with these regions,

(d) to provide a laminated tube that allows any **surface charge** that does buildup on the inner surface of the tube to be **bleed away through the metal laminates** that made up the tube,

(e) to provide a laminated tube that can transfer substantially all gas-phase ions from **ion sources that operate at pressures greater than atmospheric pressure** delivering the ions into a region at or near atmospheric pressure where they can be sampled by conventional atmospheric interfaces, either aperture or glass tube based, to mass spectrometers or other analytical devices, such as, ion mobility spectrometers,

(f) to provide a laminated tube that can transfer substantially all gas-phase ions or charged particles from **ion sources that operate at pressures greater than atmospheric pressure** delivering the ions into a region at or near atmospheric pressure where they can be directed at surfaces for deposition or surface chemistry, or reacted with other gas phase species or particulate materials.

(g) to provide static focusing or shaping of the electric fields at the **inlet** of the laminated tube, which will focus a substantial proportion of ions into the inlet of the tube irregardless of the source of ions,

(h) to provide dynamic or static focusing or shaping of the electric fields at the **exit** of the laminated tube at lower pressures, which will prevent ions from being lost due to dispersion or scattering as the ions exit the tube,

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(i) to provide dynamic focusing or shaping of the electric fields of **at least one of the multitude of conducting electrodes** to select or store ions inside the tube,

(j) to provide to the operator a **user controllable or tunable field ratios** at the entrance to the tube and along the entire length of the tube that results in improved transfer of ions from higher pressure regions into lower pressure regions,

Further objectives and advantages are to provide a laminated tube which can be easily and conveniently incorporated into existing atmospheric interfaces without the need for extensive or major reconstruction of the interface, which is simple to operate and inexpensive to manufacture, which can be used with either highly dispersive or low electrostatic or electrodynamic field ion sources; to provide a tube which can be manufactured by the techniques of microelectronics fabrication; which obviates the need for the ion source to be proximal to the inlet into the ion collection region or mass spectrometric device; etc. Still further objects and advantages will become apparent from a consideration of the ensuing descriptions and drawings.

DRAWING FIGURES

In the drawings, closely related figures have the same number but different alphabetic suffixes.

Figs 1A and 1B show computer simulation of ion trajectories at the entrance of capillary tubes where ions are transported from A) a 200 V/mm entrance region into the relatively field-free inner volume of the tube, and B) a 2000 V/mm entrance region into the relatively field-free inner volume of the tube. Note the field penetration into the tube results in significant losses due to field dispersion in the region at or near the entrance. A high ratio of outer to inner field will make the effective ion sampling aperture (a_{eff}) quite small.

Fig 2 illustrates the flow properties of gas traveling through the entrance region of a capillary tube. The initial region (Region of Flow Development) is where the gas goes from approximately plug flow to parabolic flow. In this region there is a

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significant increase in linear velocity of the gas. Under ideal conditions the gases will approach the speed-of-sound in this region.

Fig 3 is a graph of the Entrance Boundary Region (Lent) for fully developed as a function of tube diameter calculated for 25 cm tubes.

Figs 4A thru 4C show computer simulation of the ion trajectories for various operating modes of laminated tubes.

Figs 5A thru 5D shows cross-sectional illustrations of various laminated tubes.

Fig 6 shows graphs of the electric potential experienced by the charged particles or ions as they pass through a laminated tube: **Curve A** showing the potential minimizing (point b') near the entrance of a tube and showing the gradual (Section c') increase in the potential as the ions move further down the tube toward the exit of the tube, **Curve B** showing the potential minimizing (point b') near the exit of a tube and with a sharp increase in the potential as the ions move further down the tube toward the exit of the tube, **Curve C** showing the potential energy minimizing (point b') in the middle of a tube with a gradual decrease in the potential (Section a') as the ions move from the entrance of the tube to point b and a gradual increase in the potential (Section c') as the ions move from point b toward the exit of the tube.

Fig 7 shows a laminated tube configured with an atmospheric pressure ion source with funnel-well optics.

Fig 8 shows a similar laminated tube configured with a high-transmission element incorporated into an atmospheric pressure ionization source with funnel-well optics.

Fig 9 shows a similar laminated tube configured with a laminated high-transmission element incorporated into an atmospheric pressure ionization source with funnel-well optics.

Fig 10 shows a three-dimensional cutaway of the laminated tube shown in Fig 5A.

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Fig 11 shows a potential surface of the tube laminate showing the bottom of the potential well displaced from the entrance aperture where inner-tube flow is well established.

REFERENCE NUMERALS IN DRAWINGS

1	ion-source region	140	ring insulator
2	tube entrance region	150	sample source
3	tube exit region	160	delivery means
10	metal laminate or layers (designated with a through n subscripts)	170	ion-source entrance wall
12	heat source	180	ring insulator
14	high voltage supplies	190	ring insulator
18	general direction of ion flow	200	ion source gas source
20	base layers (designated with a through m subscripts)	202	ion source gas inlet
30	tube inner surface or lumen	210	tube entrance gas source
40	entrance aperture	212	tube entrance gas inlet
42	tube exhaust outlet	220	exhaust destination
44	tube exhaust destination	222	exhaust outlet
50	exit aperture	230	high-transmission element
60	exit region wall element	240	shielding electrode
62	aperture	250	back lens
70	ion-collection region	260	laminated high-transmission insulator
80	exit region wall element	262	front HTE laminate
100	entrance region wall element	264	back HTE laminate
110	electrode	266	interlaminate space
120	aperture	267	back lens
130	ion-source cylindrical wall	268	front lens
		300	equipotential lines
		302	potential well bottom

DESCRIPTION - FIGS 5A, 7 AND 10 — PREFERRED EMBODIMENT

A preferred embodiment of the laminated-tube or just tube of the present invention is illustrated in Fig 5A, Fig 7, and Fig 10. The tube is made-up of a series of thin concentric planar metal laminates or layers **10**, designated **10_a**, **10_b**, **10_c**, ... **10_{n-2}**, **10_{n-1}**, **10_n**, separated from each other by a thin base layers **20**, designated **20_a** through **20_m**, of uniform cross section and thickness consisting of non-conducting insulating material, the aggregate of metal laminate/base pairs forming a lumen, defined by a laminate/base inner surface or lumen **30**. The tube thus has an entry **40** and an exit aperture **50** for introducing gases and gas-phase ions or charged particles from a tube entrance region **2** into a tube exit region **3** where they are transported toward an ion-collection region **70**. Ions from a source region **1** and a small portion of the gases are transferred to an ion-collection region **70**, as shown by the general direction of ion flow **18**.

The collection region **70** in this embodiment is intended to be the vacuum system of a mass spectrometer (interface stages, optics, analyzer, detector), such as but not limited to quadrupole, ion traps, time-of-flight, etc.; or other low-pressure ion and particle detectors. The ion source region **1** is intended to be, but not limited to, atmospheric pressure sources of ions or charged particles; including electrospray, atmospheric pressure chemical ionization, discharge and plasma sources, photo-ionization sources, laser ionization sources, and natural and synthetic sources of ions and charged particles such as sprays.

In the preferred embodiment, the base layers **20** are glass. However the base can consist of any other material that can serve as a nonconductive insulator, such as nylon, quartz, VespelTM, ceramic, various impregnated or laminated fibrous materials, etc. Alternatively, the base can consist of other resistive or dielectric material, such as ferrite, ceramics, etc., or laminates of insulator and dielectric materials. The bases **20** may vary in both cross-section and thickness depending on the field requirements for optimal transmission and the field requirements in the tube

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entrance region **2**, the tube exit region **3**, and inside the lumen **30** of the tube. The metal laminates **10** are fabricated from a conducting and preferably inert material, such as stainless steel, brass, copper, aluminum, etc. Heat may be supplied to the tube through a heat source **12** such as heating elements (isolated) or heated gas surrounding the tube. High voltage supplies **14** supply the voltages supplied to each metal laminate **10**. Voltages can be provided to each laminate from separate supplies or through any variety of voltage divider circuits in order to deliver the precise voltage to each metal laminate from one or more voltage supplies of required voltage magnitude and polarity.

Sample from a source **150** is delivered to the ion-source region **1** by a delivery means **160** through an ion-source entrance wall **170**. The ion-source chamber **1** is bounded by the wall **170**, an electrode **110**, and an ion-source cylindrical wall **130**. The wall **170** is electrically isolated from the ion-source cylindrical wall **130** by a ring insulator **180** while a second ring insulator **190** isolates the cylindrical wall **130** from the electrode **110**. Sample from the source **150** are gas-phase ions or charged particles or, alternatively, are neutral species, which are ionized or desorbed in the ion-source chamber **1**. Heat may be applied directly to the ion-source region from the heat source **12**. Heat may also be added to the gas from an ion source gas source **200** by heating an ion source gas inlet or any variety of methods for applying heat to confined regions containing gases.

Upstream of a metal entrance region wall element **100** of the tube is a tube entrance region **2**, the electrode **110** with an aperture **120**, and an ion-source region **1** adapted to contain a gas containing gas-phase ions. The element **100**, electrode **110**, and a ring insulator **140** bound region **2**. The pressure in regions **1** and **2** should be sufficiently high to maintain viscous or chock flow through the tube, and in most applications it will be atmospheric pressure or greater. Any combination of lumen and length of the tube can be selected to limit the flow of gas from the ion-source region **1** and the tube entrance region **2** into the tube exit region **3** so that the

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pressure can be maintained at pressure difference across the tube. Excess gas in region **3** is evacuated through an exhaust outlet **222** to an exhaust destination **220**.

A DC voltage is applied to each metal laminate **10**, exit region wall elements **60**, **80**, elements or electrodes **100**, **110**, and wall **130**, **170** creating an electrical field, although one or two separate power supplies in conjunction with resistor chains can also be used to supply the desired and sufficient potential to each laminate, electrode, and element. Additional lens elements can be incorporated between wall elements **80** and **60** if desired in order to focus the ions at the exit of the tube. Alternatively, in addition to the DC potential an RF potential may be applied to each successive metal laminate **10** so that the RF voltages of each successive metal laminate is 180 degrees out of phase with the adjacent metal laminate, although other relationships for the applied RF field would be likely appropriate. Under this embodiment, an electric field is created using a power supply and a resistor chain to supply the desired and sufficient voltage to each metal laminate to create the desired potential gradient throughout the tube, and focus and confine the ions to the center of the tube.

Gas can be added for concurrent flow of gas from region **1** into region **2** from the source gas source **200** introduced through the source gas inlet **202**. In addition, gas can be added for a counter-flow of gas from region **2** into **1** from a tube entrance gas source **210** through a tube entrance gas inlet **212**. Excess gas can be exhausted through the exhaust outlet **222** toward the exhaust destination **220**. All gas supplies are regulated, metered, of adequate purity, and may be optionally heated to the meet the needs of the ion transmission application and to prevent condensation.

FIGS 8 AND 9 — ADDITIONAL EMBODIMENT

Additional embodiments of the tube are shown in Figs 8 and 9. Fig 8 shows a tube with a high-transmission element **230** with a back lens **250**, and a shielding electrode **240** (as described in our co-pending U.S. patent application, Ser. No.

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09/877,167 entitled "Apparatus and Method for Focusing Ions and Charged Particles at Atmospheric Pressure"), replacing the electrode **110**, sandwiched between the ion-source region **1** and the entrance region wall element **100** of the tube; Fig 9 shows a tube with a laminated high-transmission element **260** made up of front laminate **262**, back laminate **264**, interlaminate space **266**, back lens **267**, front lens **268**, and shielding electrode **240** (as described in our co-pending U.S. provisional patent application, Ser. No. 60/384,869 entitled "Laminated Lens for Focusing Ions from Atmospheric Pressure"), replacing the electrode **110**, sandwiched between the ion-source chamber **1** and the entrance region wall element **100**.

FIGS 5B, 5C AND 5D — ALTERNATIVE EMBODIMENTS

There are various possibilities with regard to the geometry of the lumen of the tube. Fig 5B shows a cross-sectional view of a tube composed of metal laminates with each adjacent laminate/base pair has a smaller diameter than the previous aperture, the collection of the apertures thus forming a funnel-shaped lumen. Fig 5C shows a cross-sectional view of a tube composed funnel-shaped lumen at the entrance and exit thus forming an hour glass shaped lumen. Fig 5D shows a cross-sectional view of a tube composed of laminates/base pairs with two tube diameters; the entrance diameter being larger than the exit diameter. The excess gas is exhausted mid-tube through a tube exhaust outlet **42** to a tube exhaust destination **44**.

Alternatively, the tube can be manufactured by using the techniques of micro-electro-mechanical systems commonly referred to as MEMS: photolithography for creating patterns, etching or ablation for removing material, and deposition for coating surfaces with specific materials.

The ion collection region **70** is a general description for any device that is intended for use with streams of ions or charged particles. These include mass spectrometers, ion mobility spectrometers, light scattering detectors, particle

detectors, ion deposition devices, particle deposition devices, semi-conductors fabrication devices, and printers.

ADVANTAGES

From the description above, a number of advantages of out laminated tube become evident:

(a) The delayed dispersion of ions until the tube flow is fully developed eliminates the substantial entrance losses associated with conventional tube devices.

(b) The ability to precisely control the electric field the entire length of the tube allows the tube to operate at high electric field in the entrance region and low electric field in the exit region. This allows for maximum ion transmission without electrical breakdown near the minimum of the Paschen Curve.

(c) The significant improvement in ion transmission minimizes contamination on the surfaces of the tubes and minimizes the occurrence of charging related to contamination.

(d) The use of relatively small capacitive surfaces within the laminated tube reduces the likelihood of surface charging and catastrophic discharges.

(e) The use of rf voltages on elements with fully developed flow assists in focusing the ions within the tube or at the exit and minimizes diffusion losses with longer tubes.

(f) The increased effective aperture associated with the device allows efficient collection of ion beams emanating from high compression funnel/well ion optics at atmospheric pressure.

(g) The higher ion transport efficiency of the laminated tube can also result in a much reduced gas load on the vacuum system for a given ion current. This has significant benefit in reducing the pumping requirements, cost, and complexity of vacuum systems associates with mass spectrometry.

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(h) The use of rf voltages on selected laminated elements can be used to selected ions in either high pass filter mode or band pass mode. Higher selectivity of the inlet of a mass spectrometer has the significant benefit of reducing interferences and improving analytical results.

(i) The use of above atmospheric pressure sources with high transmission current to atmospheric pressure regions presents the analyst or fabricator with a relatively high current and low field source of ions for implantation, deposition, or reaction with surfaces, particles, or gases.

(j) The use of above atmospheric pressure sources with high current will have a direct benefit as a low field external source of reagent ions for atmospheric pressure chemical ionization. This will improve sensitivity and potentially specificity of mass spectral analysis.

(k) The use of laminated tubes in parallel arrays will have the advantage of increasing the ion transmission cross-section while significantly reducing the gas load on the low-pressure side of the capillary.

OPERATION - FIGS 5 THROUGH 11

The manner of using the laminated tube to control the potential applied to the ions or charged particles entering, traversing, and exiting the tube is described. Gas-phase ions or charge particles formed in either low- or high-field sources, including, but not limited to electrospray, atmospheric pressure chemical ionization, photo-ionization, electron ionization, laser ionization and desorption (including matrix assisted), inductively coupled plasma, discharge ionization; etc. are presented to the entrance region 2 by any variety of focusing and transmitting devices, incorporating optical and aerodynamic means to collect ions at or near the entrance of the tube. The device is operated with the entrance region 2 held at a pressure substantial higher than the pressure maintained in region 3. Typical values from an atmospheric source would be atmospheric or near atmospheric in region 2 and 1 to

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10 Torr in region **3**. This pressure difference will facilitate viscous tube flow inside the tube. Under these flow conditions, the gas velocities at the entrance of the tube will be much less than the velocities inside the tube after the flow profile is fully developed. It is the intension of this device to delay the application of diverging fields until the velocity profile of tube flow is fully developed. Under fully developed gas flow, the diverging field will have a substantially lower effect on the transport of ions to the walls of the tube. The important consideration in the operation of this invention is the precise control of forces experienced by the ions and particles at each point along the pathway through the tube. The decreased loss of charged species to the walls will result in increased transmission of ions and charged particles for subsequent collection, focusing, and or detection.

Fig 6 shows the changes in the potential experienced by the ions as they move through the tube for various settings of the DC voltage of the individual metal laminates **10**. Section a' of the three graphs showing the potential experienced by the ions decreasing as they move from the entrance of the tube, through the tube, minimizing at point b'; and increasing as the ions move from point b' toward the exit of the tube, section c'. All three of these configurations illustrating that the electric potential of the ions can be controlled at any point along the tube. In this manner, the divergence of ions can be delayed until the gas flow profile within the tube is fully developed. Under these conditions, viscous flow will have a more dominant effect on the motion of ions and prevent them from migrating to the walls of the tube.

Curve A shows the change in potential of the ions as they move through the tube, the potential decreasing at a steep linear slope (section a') from the entrance of the tube to point b', with point b' near the entrance of the tube, and abruptly increasing the potential en (section c') at a swallow linear slope from point b' to the exit of the tube. Thereby gradually exposing the ions to the dispersive electric fields (section c') of the increasing potentials on the metal laminates.

Curve B shows the change in potential of the ions as they move through the

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tube, the potential energy decreasing at a shallow linear slope (section a') from the entrance of the tube to point b', with point b' near the exit of the tube, and increasing (section c') at a steep linear slope from point b' to the exit of the tube. Thereby allowing the ions to be well established in the center of the tube before abruptly increasing the potential of the ions and exposing the ions to the dispersive electrical fields (section c') of the increasing potential on the metal laminates.

Curve C shows the changes in the potential of the ions as they move through the tube, the potential gradually decreasing (section a') from the entrance of the tube to point b', in the middle of the tube, and then gradually increasing the potential of the ions (section c') to the exit of the tube. Thereby allowing gradual changes in the potential of the ions as they pass through the tube and exposing the ions to focusing (section a') and dispersive (section c') electrical field lines gradually.

The tube-laminate allows the matching of the flow profile and the electric fields experienced by the ions and particles as they traverse the tube. Tube flow can be controlled by tube diameter, pressure difference across the tube, entrance and exit geometry, gas composition, temperature, and other surface properties (some variations are illustrated in Figs 5A-D). The device will operate by selecting the "experiment-required" flow parameters, then matching the fields to the application. For example, placing the bottom of the potential well within the tube will allow the ion source to operate at or near ground potential, while allowing the exit of the tube to also be held at or near ground potential. The potential gradient can also be precisely controlled with this device in order to minimize high field at the lower pressure end of the tube; thus preventing electrical discharge when the exit of the tube is under vacuum.

Fig. 11 shows a potential surface of the tube-laminate in operation with the ion motion **18** being perpendicular to equipotential lines **300** within the tube. At the bottom of a potential well **302** the ions cease to follow the diverging field because their motion is dominated by viscous flow.

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The electric potential experienced by ions traveling through the tube is governed dimensionally by the diameter, spacing, and applied voltage of the metal laminates **10**. Although the enclosed figures show uniform spacing between respective laminates, the distance between each laminate, controlled by the specific dimension of base layers **20** can vary from layer to layer depending on the local field requirements. Large distances along the tube with uniform field requirements can be accommodated with a single base layer.

Another important mode of operation of the tube-laminate is from above atmospheric pressure sources of ions into at (or near) atmospheric pressure exit region **3**. Note that tube flow is governed by pressure difference, not absolute pressure. The tube-laminates can also have applications in series with tubes and multiple potential wells.

When the tube laminate is operated with vacuum conditions in region **3**, any number of evacuation devices can serve as the exhaust destination; including, roughing pumps, turbo pumps, cryo-pumps, etc.

Gas flowing in a direction that is counter to the movement of ions will serve to reduce or eliminate contamination from particulate materials and neutral gases in the tube. Operation with a counter-flow of gas is accomplished by adding a sufficient flow of gas (optionally heated) from the entrance tube gas source **210** flowing out through the inlet **212**, through the aperture **120** and into the ion-source region **1**, to prevent contamination of the wall element **100** and prevent droplets from entering the entrance aperture **40** of the tube. In addition, lower mobility charged particles or ions may also be swept away in the counter-flow of gas.

CONCLUSION, RAMIFICATION, AND SCOPE

Accordingly, the reader will see that the laminated tubes of this invention can be used to transport ions from an atmospheric or higher pressure ion source region into lower pressure regions. Further more, the laminated tube has the additional

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advantages in that:

- It is compatible technology for implementation on most existing atmospheric pressure sources used in modern mass spectrometry and ion mobility spectroscopy.
- The need for high current sources of ions across pressure regimes is evident in the manufacture of semi-conductors, micro-electronics, nano-components, thin film deposition, etc.

Although the description above contains many specifications, these should not be construed as limiting the scope of the invention but as merely providing illustrations of some of the presently preferred embodiments of this invention. For example the lumen of the tube can have other shapes, such as oval, square, triangular, etc.; ions can be formed by natural or synthetic means; the number of and distance between adjacent metal laminates of the tube can vary depending on the source of ions, the ion collection region, the respective pressure of each region or a combination thereof; the laminates in the area of the inlet of the tube can have larger openings to accommodate dispersive ion sources, while the laminates at the exit of the tube can have larger openings to facilitate the formation of a an ion-beam or a combination thereof utilized; etc.

We also envision that the present device may operate with bundles of parallel tube-laminates for some ion transfer applications where smaller tube diameters or larger flows may be required. Alternatively, we envision splitting the flow (As shown in Fig 5D) in applications where minimal conductance into vacuum is required.

We also envision the use of both dc and rf voltages within the lumen of this device to selectively manipulate ions from any variety of sources to any variety of ion destinations.

Thus the scope of the invention should be determined by the appended claims and their legal equivalents, rather than by the examples given.